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(54) Title: **High shock resistant thermoplastic resin composition**

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Text of specifications

1. Title

High shock resistant thermoplastic resin composition.

2. Claims

A high shock resistant thermoplastic resin composition prepared by compounding a graft copolymer (I) with at least one type of thermoplastic resin (II) selected from styrene resin, methyl methacrylate resin vinyl chloride resin, acrylonitrile-styrene resin and polycarbonate resin in such a way that the content of acrylic rubber (A) in the final resin composition is 3-40 wt. %.

The graft copolymer (I) is prepared by polymerisation of 93-30 parts by weight

of grafting monomer(s) that consists of 30^A-100 wt.% of at least one type of monomer selected from styrene, acrylonitrile and methyl methacrylate and 0-30^A wt% of at least one other monomer that is copolymerisable with this monomer(s) and has the CH₂=C< group, in the presence of 7-70 parts by weight (in terms of solid content) of a swollen rubber latex.

The swollen rubber latex is prepared by adding 0.1-5 parts by weight (in terms of the solid content) of a specific acid-containing copolymer (B) latex to 100 parts by weight (in terms of solid content) of an acrylic rubber (A) latex.

Here, the acrylic rubber (A) latex is prepared by polymerisation of 100-50 wt.% of an alkyl acrylate (i) with 2-10 carbons in the alkyl group, 0-50 wt.% of a monofunctional monomer (ii) copolymerisable with (i) and 0-5 wt.% of a multifunctional monomer (iii) that is copolymerisable with (i) and (ii).

The acid-containing copolymer (B) latex is prepared from a monomer component (c) comprising 3-30 wt.% of at least one unsaturated acid (iv) selected from acrylic acid, methacrylic acid, itaconic acid and crotonic acid, 97-35 wt.% of at least one type of alkyl acrylate (v) having 1-12 carbons in the alkyl group and 0-48 wt.% of at least one other monomer (vi) copolymerisable with the monomers (iv) and (v). The copolymer (B) is prepared by polymerising 5-90 wt.% of the monomer component (c) excluding the unsaturated acid component (iv), in the first step, and then polymerising the remaining 95-10 wt.% of the monomer component (c) containing the unsaturated acid(s) (iv).

3. Detailed description of the invention

The present invention deals with a new type of highly shock resistant thermoplastic resin composition.

High shock resistant resins, typical of which are ABS resins and high impact polystyrene are prepared by graft polymerisation of styrene, acrylonitrile, methyl methacrylate and other such monomers to a rubber component. The composition and structure of the graft copolymer, the rubber content, the method of polymerisation, etc have significant bearing on the physical properties of the final resin composition. Especially when the rubber component is graft polymerised by emulsion polymerisation, the particle size of the rubber that forms the substrate determines the shock resistance and workability of the final product, as is well-known. The greater the particle size of the rubber, the better are the impact resistance and workability of the resin.

Because of this, several attempts have been made to make the rubber particles as large as possible. Various suggestions have been put forth.

The present applicant also had earlier suggested a high shock resistance resin composition prepared by polymerisation of at least one type of monomer selected from styrene, acrylonitrile, and methyl methacrylate, in the presence of swollen rubber latex. This suggestion was based on the finding that particles of diene rubber and acrylic rubber could be swollen by the use of a copolymer latex prepared from an unsaturated acid monomer and an alkyl acrylate. Further studies revealed that swollen rubber particles with better stability could be created if a small amount of an acid radical-

containing copolymer latex, which consists of an unsaturated acid and an alkyl acrylate and has a specific double layer structure, is added to acrylic rubber latex. It was also found that the final resin composition had superior characteristics if a swollen rubber latex of this type was graft polymerised with least one monomer selected from styrene, acrylonitrile and methyl methacrylate and a specific thermoplastic resin is compounded with it to the extent that the acrylic rubber content in the resin composition remains within a certain range. This led to the new invention.

In short, the new invention is a high shock resistant thermoplastic resin composition prepared by compounding a graft copolymer (I) with at least one type of thermoplastic resin (II) selected from styrene resin, methyl methacrylate resin vinyl chloride resin, acrylonitrile-styrene resin and polycarbonate resin in such a way that the content of acrylic rubber (A) in the final resin composition is 3-40 wt.%.

The graft copolymer (I) is prepared by polymerisation of 93-30 parts by weight of grafting monomer(s) that consists of 30-100 wt.% of at least one monomer selected from styrene, acrylonitrile and methyl methacrylate and 0-30 wt% of at least one other monomer that is copolymerisable with this monomer(s) and has the $\text{CH}_2=\text{C}<$ group, in the presence of 7-70 parts by weight (in terms of solid content) of a swollen rubber latex.

The swollen rubber latex is prepared by adding 0.1-5 parts by weight (in terms of the solid content) of a specific acid-containing copolymer (B) latex to 100 parts by weight (in terms of solid content) of an acrylic rubber (A) latex.

Here, the acrylic rubber (A) latex is prepared by polymerisation of 100-50 wt.%

of an alkyl acrylate (i) with 2-10 carbons in the alkyl group, 0-50 wt.% of a monofunctional monomer (ii) copolymerisable with (i) and 0-5 wt.% of a multifunctional monomer (iii) that is copolymerisable with (i) and (ii).

The acid-containing copolymer (B) latex is prepared from a monomer component (c) comprising 3-30 wt.% of at least one unsaturated acid (iv) selected from acrylic acid, methacrylic acid, itaconic acid and crotonic acid, 97-35 wt.% of at least one type of alkyl acrylate (v) having 1-12 carbons in the alkyl group and 0-48 wt.% of at least one other monomer (vi) copolymerisable with the monomers (iv) and (v). The copolymer (B) is prepared by polymerising 5-90 wt.% of the monomer component (c) (not containing the unsaturated acid component) in the first step, and then polymerising the remaining 95-10 wt.% of the monomer component (c) containing the unsaturated acid.

The acrylic rubber (A) of the new invention can be a homopolymer of an alkyl acrylate with 2-10 carbon atoms in the alkyl group, a copolymer consisting of 50 wt.% or more of the alkyl acrylate and 50 wt.% or less of a monofunctional monomer that is copolymerisable with this alkyl acrylate, or a copolymer in which a multifunctional monomer is also used to the extent of not more than 5 wt.%. Such an acrylic rubber can be easily prepared by a standard method of emulsion polymerisation. The structure of the acrylic rubber (A) is not specified. It can have a uniform structure or a multi-layered structure in which the interior of the rubber particles consists of a cross-linked hard resin or a rubber with a different degree of cross-linking. The catalyst, emulsifier, etc used at the time of production are also not specified. The particle size of the rubber component (A) is 0.04-0.2 μ .

In the newly invented method, the acid-containing copolymer (B) latex is used as a swelling agent to swell the acrylic rubber (A) in the latex and it is essential for (B) to be in the form of a latex. The acid-containing copolymer (B) latex is prepared from a monomer component (c) comprising 3-30 wt.% of at least one unsaturated acid (iv) selected from acrylic acid, methacrylic acid, itaconic acid and crotonic acid, 97-35 wt.% of at least one type of alkyl acrylate (v) having 1-12 carbons in the alkyl group and 0-48 wt.% of at least one other monomer (vi) copolymerisable with the monomers (iv) and (v). The copolymer (B) is prepared by polymerising 5-90 wt.% of the monomer component (c) (not containing the unsaturated acid component) in the first step, and then polymerising the remaining 95-10 wt.% of the monomer component (c) containing the unsaturated acid. It is essential for the copolymer (B) to have a two-layer structure and to be present in the form of a latex. The size of the copolymer (B) particles in the latex significantly influences its swelling capacity and the preferable particle size range, from a practical point of view, is 0.05-0.2 μ .

The above-specified unsaturated acid (iv), which is a component of the acid-containing copolymer (B), can be a single acid or a mixture of more than one acid.

The content of unsaturated acid(s) in the copolymer (B) is 3-30 wt.%. With less than 3 wt.%, the copolymer (B) would not have sufficient swelling capability, whereas with 30 wt.% its swelling capacity becomes too high resulting in the formation of particles that are too large, exceeding 1 μ , which is not suitable.

The optimum content of the unsaturated acid(s) depends on the hydrophilic property of the acrylate used. If the acrylate is highly hydrophilic, the desired swelling can be achieved with a relatively small amount of unsaturated acid. But if the unsaturated acid content is large when the acrylate is highly hydrophilic, this can lead to undesirable collapse of the latex. On the other hand, when the acrylate is not very hydrophilic, the desired swelling cannot be achieved unless the unsaturated acid content is on the higher side of the specified range. For example, when the acid-containing copolymer (B) latex particles have a single layer structure, an unsaturated acid content of 5-10 wt.% is optimum if the acrylate is highly hydrophilic, such as methyl acrylate or ethyl acrylate. But if it is a rather hydrophobic alkyl acrylate with 4 or more carbons in the alkyl group, such as butyl acrylate or 2-ethylhexyl acrylate, the optimum unsaturated acid content is 13-20 wt.%. With highly hydrophilic acrylates, the system easily becomes unstable even when the unsaturated acid content is 5-10 wt.%, often producing a cullet of coarse particles. But with hydrophobic acrylates of the type mentioned above, the system is more stable and uniformly swollen particles can usually be obtained. The alkyl acrylate, which is a component of the acid-containing copolymer (B), must have 1-12 carbons in the alkyl group. It can be a single alkyl acrylate or a mixture of more than one. The alkyl acrylate content of the copolymer (B) is 97-35 wt.%. If it is outside this range, the effect of the new invention will not be achieved satisfactorily. If other monomers like methacrylate, styrene and acrylonitrile are used instead of the alkyl acrylate, the desired effect cannot be achieved at all. However, it is possible to replace up to 50% of the required amount of alkyl acrylate with such other monomers.

Examples of other copolymerisable monomers that may be used, if needed, for preparing the acid-containing copolymer(B) are unsaturated aromatic compounds like styrene, α -methyl styrene, and vinyl toluene; unsaturated nitrile compounds like acrylonitrile, methacrylonitrile; and alkyl(metha)acrylates with 1-12 carbons in the alkyl group. These can be used singly or in combinations. The suitable amount of such copolymerisable monomers is 0-48 wt.% of copolymer (B). If this exceeds 48 wt.%, the effect of the new invention cannot be achieved.

When we compare the acid-containing copolymer (B) latex having the above-mentioned two-layer structure with other copolymer latexes having the same monomer composition but with single layer structure, prepared by copolymerising in one step, we can see that the copolymer (B) of the new invention, which is designed to have a higher unsaturated acid content in the outer layer of the latex particles, even though the total acid content is the same, has much greater swelling effect on the acrylic rubber particles. Because of this, swollen rubber of larger particle size can be produced in a short time. Moreover, the swollen rubber latex made in this manner has better stability with less change in particle size with the passage of time.

The suitable amount of the acid group-containing copolymer (B) latex to be added is 0.1-5 parts by weight, most preferably 0.5-3 parts by weight, for 100 parts by weight (all in terms of the solid content) of the acrylic rubber (A) latex.

If necessary, 0.05-4 parts by weight of an inorganic electrolyte may be added for 100 parts by weight (in terms of the solid content) of the acrylic rubber latex to

make the swelling even more effective and to improve the stability of the swollen rubber latex.

The suitable electrolytes are ordinary inorganic salts like KCl, NaCl and Na_2SO_4 .

These inorganic electrolytes can be pre-mixed with the acrylic rubber latex at the time of its polymerisation, which gives an equally good effect as addition at the time of swelling of the rubber.

The pH of the acrylic rubber (A) latex needs to be maintained at 7 or higher at the time of the swelling treatment in the newly invented method. If the pH is in the acidic range, the addition of the acid group-containing copolymer (B) latex would not have the desired effect and the resin composition aimed at by the new invention cannot be produced effectively.

For example, butyl polyacrylate rubber latex polymerised using sodium octyl sulphosuccinate as the emulsifier and sodium persulphate as the catalyst has a pH of 2-3. Under this condition, absolutely no swelling occurs even if the acid-containing copolymer latex is added. But, if its pH is adjusted to 7 or higher beforehand, by adding a small amount of an alkali like potassium hydroxide, the rubber can be easily swollen to a large particle size.

Adjusting the pH of the acrylic rubber (A) latex to 7 or higher can be done during the polymerisation of the acrylic rubber or just before the swelling treatment.

In the next step, 93-30 parts by weight of grafting monomer(s) are polymerised in the presence of 7-70 parts by weight of the swollen rubber latex prepared as described above, to obtain graft copolymer (I) used in preparing the newly invented high shock resistance thermoplastic resin. The grafting monomer(s) consists of 30-100 wt.% of at least one monomer selected from styrene, acrylonitrile or methyl methacrylate and 0-30 wt.% of at least one monomer copolymerisable with this monomer and having the $\text{CH}_2=\text{C}<$ group.

Styrene, acrylonitrile and methyl methacrylate are used singly or in combinations to the extent of 30-100 wt.% in the grafting monomer(s).

The monomers that are copolymerisable with the above monomers and having the $\text{CH}_2=\text{C}<$ group are aromatic vinyl compounds such as styrene, α -methyl styrene and vinyl toluene; unsaturated nitrile compounds like acrylonitrile and methacrylonitrile; and alkyl(metha)acrylates with 1-12 carbons in the alkyl group. These are used singly or in combinations. Their content in the total monomers for grafting is 30 wt.% or less. It is unsuitable to use these alone as the monomers for grafting.

Practical examples of grafting monomer mixtures are styrene-acrylonitrile, styrene-alkyl acrylate, acrylonitrile-methyl methacrylate, methyl methacrylate-alkyl acrylate and acrylonitrile-alkyl acrylate. Mixtures containing three or more monomers of this type can also be used.

Commonly used emulsifiers and catalysts are used for this emulsion graft polymerisation. Their types and amounts are not specified in this invention.

In the graft polymerisation on to the swollen rubber, the grafting monomer(s) may be added in one go, in batches, or in a continuous manner. Alternatively, the monomers may be graft polymerised sequentially one after another.

The graft polymer (I) prepared in this manner and at least one type of thermoplastic resin (II) selected from styrene resin, methyl methacrylate resin, vinyl chloride resin, acrylonitrile-styrene resin and polycarbonate resin are compounded in such a way that the content of the acrylic rubber (A) in the final resin composition is 3-40 wt.%, to prepare a high shock resistance resin composition.

Here, the “styrene resin” means a homopolymer or copolymer having 50 wt.% or more of styrene units. Similarly, “methyl methacrylate resin” means a homopolymer or copolymer having 50 wt.% or more of methyl methacrylate units and “vinyl chloride resin” means a homopolymer or copolymer having 50 wt.% or more of vinyl chloride units. The “acrylonitrile-styrene resin” includes acrylonitrile- α -methyl styrene copolymer and acrylonitrile-styrene- α -methyl styrene copolymer. “Polycarbonate resin” means a homopolymer or copolymer having 50 wt.% or more of polycarbonate units.

In the new invention, the graft polymer (I) and the thermoplastic resin (II) may be mixed together while they are still in emulsion form if the resin (II) has been prepared by emulsion polymerisation. Otherwise, they may be mixed in the form of

powders or powder and beads using a Henschel mixer, extruder, Banbury mixer, heated rollers, or any other suitable device.

Antioxidants, lubricants, colouring agents, fillers, etc may be added to the newly invented resin composition.

The newly invented resin composition has good weather resistance and impact resistance, besides having a very good appearance when made into injection-moulded products. Therefore, it has significant industrial value.

The new invention is explained below in greater detail referring to some practical examples of its application. In these examples, “parts” and “%” respectively mean “parts by weight” and “wt.%”.

Example 1

I. Preparation of acrylate ester rubber (A – 1)

The mixture of the materials listed below was placed in a glass flask and the atmosphere was replaced with nitrogen while stirring the contents.

Deionised water	200 parts
Potassium oleate	3 “
n-Butyl acrylate	100 “
Triaryl isocyanurate	0.5 “

After heating this mixture to 50° C, 0.3 parts of potassium persulphate was added to initiate the polymerisation. The reaction was allowed to continue at 50° C for 4 h and another 2 h at 70° C. The per cent reaction was 99, the mean particle size of the latex obtained was 0.09 μ and the pH was 9.1.

II. Synthesis of acid-containing copolymer (B - 1) used as swelling agent

n-Butyl acrylate	75 parts
Potassium oleate	2 “
Sodium dioctyl sulphosuccinate	1 “
Cumene hydroperoxide	0.3 “
Sodium formaldehyde sulfoxylate	0.3 “
Water	200 “

The above mixture was polymerised for 1.5 h at 70° C. Following this, a mixture of the materials listed below was added to it drop-wise at 70° C, taking a total of 1 h. This was followed by 1 h of stirring. The reaction yielded a copolymer latex with particle size 0.12 μ . The per cent reaction was 98.

III. Preparation of swollen latex

100 parts (in terms of the solid content) of the A - 1 latex was placed in a container. While stirring it, 2.0 parts (in terms of solid) of the B - 1 latex was added at room temperature and stirred for 30 minutes. The latex was sampled after this stirring, and also after letting it stand for 5 days, to determine the mean particle size. The results are given in Table 1.

IV. Synthesis of graft polymer (G - 1)

Graft polymerisation was carried out at 80° C for 4 h using the swollen latex immediately after 30 minutes of stirring and the components listed below, to synthesize the graft polymer.

Swollen rubber latex (in terms of solid)	60 parts
Methyl methacrylate	8 “
Acrylonitrile	8 “
Styrene	24 “
n-Octyl mercaptan	0.04 “
Potassium persulphate	0.2 “
Potassium oleate	1.0 “

V. Evaluation of physical properties

50 parts of the graft polymer obtained in Step II above and 50 parts of suspended particles prepared separately using a 20/20/60 monomer mixture of methyl methacrylate/ acrylonitrile/styrene were blended to make the content of acrylic rubber in the total resin composition 30%. After adding 1 part of barium stearate and 0.1 part of Tinuvin P (UV absorber, Ciba Geigy) the resin composition was pelletised using an extruder. These pellets were then used to make different test pieces by injection moulding and the physical properties of these test pieces were evaluated. Table 1 gives the results of the evaluation. In Table 1, the notched Izod strength was determined as per ASTM-D-256. The melt index (MI) was expressed in terms of grams of polymer that came out in 10 min at 200° C under a load of 5 kg. The surface gloss was measured by moulding 1/8th inch flat sheets. The specular gloss was determined as per ASTM-D523-62T at an angle of incidence of 60° . The same methods of evaluation were used in the subsequent examples and comparative examples also.

Comparative Example 1

For the sake of comparison, a copolymer latex (B - 2) used as swelling agent was prepared by polymerising the materials listed below in one step and the resin and mouldings were prepared using it in place of B - 1, following the methods of Example 1.

The physical properties are given in Table 1.

n-Butyl acrylate	90 parts
Methacrylic acid	10 “
Potassium oleate	2 “
Sodium dioctyl sulphosuccinate	1 “
Cumene hydroperoxide	0.4 “
Sodium formaldehyde sulfoxylate	0.3 “
Water	200 “

Comparative Example 2

A similar experiment was carried out using the acrylic acid ester rubber (A - 1) as such without swelling for preparing the graft copolymer. Test pieces were prepared as in Example 1 and their physical properties determined. The results are given in Table 1.

It can be seen from Table 1 that if the acrylic rubber (A - I) is swollen, both the impact resistance and melt index were better compared to when no swelling treatment was given to the rubber. Moreover, the newly invented type of swelling agent with double layer structure had a better swelling effect, yielding larger swollen rubber

particles in a short time, compared to a swelling agent with uniform single layer structure.

Table 1

	Rubber particle size (μ)			Notched Izod strength (kg cm/cm ²)	Melt index (g/10 min)	Surface gloss
	Initial size	Size after swelling				
		After 30 min	After 5 days			
Example 1	0.09	0.30	0.31	29	1.3	94
Comparative Example 1	0.09	0.12	0.15	13	1.0	93
Comparative Example 2	0.09	-	-	8	0.7	93

Example 2

Latex (B - 3), an acid-containing copolymer latex, was prepared using the same method as for latex (B - 1) of Example 1. However, 45 parts of butyl acrylate and 5 parts of methyl methacrylate were used in the first step and 30 parts of butyl acrylate and 20 parts of methacrylic acid were used in the second step. The rest was the same as for latex (B - 1) of Example 1. The particle size of the latex (B - 3) was 0.09 μ .

The latex (A - 1) was swollen using this latex (B - 3) in the same manner as in Step III of Example 1. Graft polymer (G - 2) was then prepared by polymerising the materials listed below, following the method of Step IV of Example 1. The swollen rubber particles had a size of 0.30 μ .

Swollen rubber (solid basis)	60 parts
Methyl methacrylate	40 “
n-Octyl mercaptan	0.04 “

Potassium persulphate

0.2 “

Separately, suspension polymerisation was carried out using methyl methacrylate. 50 parts of the beads obtained, 50 parts of the graft polymer (G - 2) and 0.3 parts of barium stearate were blended and pelletized using an extruder. After that, the physical properties of test pieces were evaluated following the method of Step V of Example 1. The samples had a notched Izod strength of 16 kg cm/cm² and a melt index of 0.4.

Example 3

20 parts of graft polymer (G - 2), vinyl chloride resin^B (degree of polymerisation 950), 3 parts of dibutyl tin maleate, 1 part of barium stearate and 0.5 part of stearic acid were blended in a Henschel mixer. Test pieces were prepared with this material by pressure moulding and their impact strength measured. The notched Izod strength was 32 kg cm/cm²

Example 4

Te acrylic acid ester rubber latex (A - 1) was swollen using an acid group-containing copolymer latex (B - I) as in Step III of Example 1. After that, it was subjected to continuous drip polymerisation using the materials listed below to prepare graft polymer (G - 3).

Swollen rubber (solid basis)	60 parts
Acrylonitrile	10 “
Styrene	30 “
n-Octyl mercaptan	0.02 “

50 parts of this graft polymer and 50 parts of suspended particles prepared separately using a 25/25(%) monomer mixture of acrylonitrile and styrene were blended to make the content of acrylic rubber in the final resin composition 30%. In addition to this, 0.5 part of barium stearate was added to this resin composition and it was pelletised using an extruder. Test pieces were prepared from these pellets by injection moulding and their physical properties evaluated. The samples had a notched Izod strength of 33 kg cm/cm² and a melt index of 1.1.

Example 5

20 parts of graft polymer (G - 3), 60 parts of polycarbonate resin, 20 parts of acrylonitrile-styrene resin and 0.5 parts of barium stearate were blended and the properties of samples prepared from the resin composition were evaluated following the methods used in Example 1. The samples had a notched Izod strength of 56 kg cm/cm² and a melt index of 4.5 under a 5 kg load at 230° C.

Example 6

30 parts of graft polymer (G - 3), 70 parts of polystyrene resin and 0.5 parts of barium stearate were blended and the properties of samples prepared from the resin composition were evaluated following the methods used in Example 1. The samples had a notched Izod strength of 90 kg cm/cm² and a melt index of 1.6.

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Translator's Notes:

1. *Please check parts marked in orange. Their translation is uncertain.*
2. *Parts where there is an apparent error in the source text are marked in pink.*

^A*This does not add up to 100. This is given as 30 wt.% throughout the text and it is rendered as such in this translation (page 2).*

^B*The amount of vinyl chloride resin is not given (page 18).*

3. *We have introduced abbreviated names such (i), (ii)...(vi), (c) etc for improving clarity and reducing repetition. We have also broken up very long sentences in the source text into smaller ones.*